Formidlingstekst - Kasper Mackeprang

Climate models are one of the most important aspects of climate research, as the state of the world and the future state of the world are predicted by these models. These models are used to take political decisions associated with the climate and it is important that these models are accurate. In this context, we have in our research group studied so-called hydrogen bonded complexes. The interest in hydrogen bonded complexes and clusters has grown in recent years, as their impact on the climate has become evident.[1–10] Hydrogen bonded complexes and clusters are systems of two or more molecules that are bonded together through hydrogen bonds. Hydrogen bonds are a specific type of interaction between molecules that are very strong and essential for life as we know it, e.g. they are responsible for the helix structure of DNA and the unique properties of water. An example of the importance of these hydrogen bonded complexes is that for a long time, the observed solar radiation on Earth could not be predicted by the existing theoretical models. It was discovered that the discrepancy between the models and the observed radiation could be attributed to hydrogen bonded complexes. Especially, some of the effects arising from the formation of small hydrogen bonded complexes containing water molecules were not considered in the previous models.[2,11–14] Upon formation of these hydrogen bonded complexes, the ability of the molecules to absorb the incoming solar radiation changes significantly.[1,2,4,5] Taking these effects into account in climate models, scientists have significantly improved the quality of the models in recent years.

Hydrogen bonded complexes and clusters are essential to the formation of atmospheric aerosols. Aerosols are fine particles in the air, and they play a central role in climate modelling, but they are associated with some of the largest uncertainties in current climate models.[15] Aerosols can be emitted directly to the Earth's

atmosphere through various processes (primary aerosols),[16] but they can also be formed through so-called nucleation processes in the atmosphere (secondary aerosols). It is particularly interesting to understand how these secondary aerosols are formed in the atmosphere in order to take their impact into account in climate models. Many of the proposed mechanisms for forming precursors (i.e. the initial systems that lead) to these secondary aerosols involve small clusters containing sulfuric acid, water and some component that stabilize the cluster through hydrogen bonds.[17] Various stabilizing components and mechanisms have been suggested.[18,18–26] For this reason, the formation and stability of small hydrogen bonded complexes and clusters are interesting, but these properties are inherently difficult to obtain.

The experimental challenge associated with the determination of formation and stability of these complexes are that the amount of complex formed is often very difficult to measure. The reason for this is that often there is far less of the complex relative to other species present in the experiment. You can compare this to a concert with 50.000 people. If 100 extra people shows up, you would not notice it, if you were standing at the stage looking over the entire audience. In our research group, we[27–36] and others[37–39] have utilized a method that was recently discovered to get around this problem and determine the formation and stability of a large series of bimolecular hydrogen bonded complexes. A bimolecular hydrogen bonded complex is a complex formed between two molecules through a hydrogen bond. The method utilizes the fact that the interaction of these complexes with light changes significantly upon the formation of the complex. If we go back to the concert metaphor, it would be like if the 100 people that showed up to the concert brought a lighter. Then it would be significantly easier to detect these people and maybe even determine how many there where. As such, we are not trying to count the amount of complex formed (count the people that showed up to the

concert), we are instead looking at their interaction with light (how many lighters we see). These types of experiments are called spectroscopic experiments, and in our specific case we are looking at what is called vibrational spectroscopy. In our experiments, we can only see the total interaction of the complexes with light (the total amount of light coming from the lighters of the people at the concert) and not the interaction from the individual complexes (not the individual lighters). Fortunately, the interaction for each complex with light is the same and in order to determine the amount of complex present, we need to know the interaction of a single complex with light. Unfortunately, for our complexes this value is often unknown and specific for each complex, but there is a solution to this.

In modern chemistry, computational chemistry is often utilized to determine or rather estimate specific chemical properties using mathematical models. Numerous models exists and they all have assumptions and approximations, and correspondingly all the calculated properties are inherently associated with uncertainties. The effectiveness of the models are often a question of accuracy *versus* speed. The calculations can be performed in seconds, minutes, hours, days, weeks, months or years depending on the model and the underlying assumptions and approximations. The accuracy and speed are generally linked such that a model that ensures high accuracy is very slow to perform. A challenge for computational chemists is, thus, to develop models that are fast but still captures the fundamental chemistry and physics behind the problem it tries to describe in order to produce accurate results. If we return to our specific problem concerning the bimolecular hydrogen bonded complex, previous models have been unable to accurately determine the ability of these complexes to interact with light. These previous models are too crude and inaccurate, they take to long to perform or they simply yield unphysical results. As such, we decided to develop a new model that captures the correct chemistry and physics of our systems and properly describe

the ability of our systems to interact with light while still being feasible to perform.

The development of this model, which we have called the Local Mode Perturbation Theory model,[40,41] has been the main focus of the Ph.D. project. It was successfully developed and implemented, and we have tested it on systems for which experimentally measured values exist. We found that it agrees very well with existing studies, however, we have also used it to question previously measured values in the literature. Additionally, we have also applied it to a large series of complexes that we have studied experimentally, and as a consequence greatly improved the results obtained from these studies. With our results and the development of this model, we are able to obtain more accurate values related to the formation and stability of these complexes. This should help our general understanding of the formation and stability of hydrogen bonded complexes, which potentially could lead to an improved understanding of the formation of aerosols and current climate models.

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